Carbon Black Filled Silicone as a Compliant Thermoelectric Material

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ABSTRACT: A compliant silicone–matrix composite containing carbon black and exhibiting volume electrical resistivity 2.3 Ω cm was found to exhibit absolute thermoelectric power +2 μV/°C.

KEY WORDS: composite, silicone, carbon black, thermoelectric, electrical, Seebeck.

INTRODUCTION

THERMOELECTRIC BEHAVIOR PERTAINS to the conversion between thermal and electrical energy. In particular, the Seebeck effect is a thermoelectric effect in which a voltage results from a temperature gradient, which causes the movement of charge carriers from the hot point to the cold point. This voltage (Seebeck voltage) is useful for temperature sensing and pertains also to the generation of electrical energy. The negative of the change in Seebeck voltage (hot minus cold) per degree C temperature rise (hot minus cold) is called the thermoelectric power, the thermopower, or the Seebeck coefficient.

The thermal stress between a thermoelectric cell and the wall of a heat exchange of a thermoelectric energy conversion system affects the thermal coupling as well as the durability. To reduce the thermal stress, compliant pads are used at the interface, although the pad acts as a barrier against thermal conduction [1,2]. If the thermoelectric material is itself compliant, a compliant pad will not be necessary.

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Metals are in general more compliant than semiconductors, but their Seebeck effect is relatively weak and they tend to suffer from corrosion. Polymers can be more compliant than metals, but they are usually electrically insulating and do not exhibit the Seebeck effect. On the other hand, a polymer contining an electrically conductive discontinuous filler (e.g., carbon black) above the percolation threshold is conductive. By using a polymer which is an elastomer (e.g., silicone), the resulting composite is resilient and compliant. Although there has been much work on the electrical conductivity of filled polymers [3–5], there has been no previous work (other than that related to conducting polymers [6,7]) on the thermoelectric behavior. This work is thus focused on the thermoelectric behavior of carbon black filled silicone [3–5].

Electrically conductive polymer-matrix composites such as carbon black filled silicone are used for electromagnetic interference (EMI) shielding and for electrostatic discharge protection. The resilience is important for the use as EMI gaskets. Due to the heating associated with the operation of microelectronics, which require shielding, the shielding material (particularly that associated with a mixed signal module) may encounter a temperature gradient. The thermoelectric effect of the shielding material would result in a voltage, which may affect the performance of the microelectronics. For example, the electrical grounding may be affected. Therefore, investigation of the thermoelectric behavior of shielding materials is desirable.

The thermoelectric power has been previously reported for flexible graphite [8], kish graphite [9,10], highly oriented pyrolytic graphite [9,11], carbon fibers [12,13] and graphite intercalation compounds [14,15]. For carbons that have not been intercalated, both positive and negative values of the absolute thermoelectric power at room temperature have been reported. The value depends on the crystallinity, defects, phonons and impurities, in addition to depending on the carrier type and concentration.

EXPERIMENTAL METHODS

The composite studied had a methylphenyl silicone matrix and a carbon black (25 ± 3 wt%) filler. The composite was COHRlastic EC-102 provided by St. Gobain, New Haven, CT. The volume resistivity of the composite was $5\,\Omega\,\text{cm}$. The composite was in the form of an isotropic sheet of thickness 1.6 mm.

The thermoelectric behavior in the through-thickness direction of the composite was investigated using the following method. A specimen of size 36 × 36 × 1.6 mm was placed with its 36 × 36 mm side on an insulator-lined hot plate. The lining was a glass fiber reinforced teflon film. Thus, a temperature gradient was generated in the through-thickness direction of the specimen. The voltage difference between the top and bottom surfaces of the specimen was measured by using electrical contacts in the form of silver paint in conjunction with copper wire. The temperatures of the top and bottom surfaces of the specimen were simultaneously measured by using two T-type thermocouples. The temperature of the hot side ranged from 24 to 70°C; that of the cold side ranged from 24 to 38°C. The temperature difference was up to 32°C. The cold side was cooled by using air, which was blown through a steel pipe (electrically insulated from the specimen by using a lining) in the direction perpendicular to the specimen surface. The heating rate of the hot side was 0.30°C/min. The cooling rate of the hot side was 0.28°C/min. Two specimens were tested.

The DC volume electrical resistivity in the through-thickness direction was measured by using the four-probe method, using silver paint in conjunction with tin-coated copper wire strand as electrical contacts. In each of the two opposite large faces of a specimen of size 36 × 36 × 1.6 mm, a current contact in the form of a loop and a voltage contact in the form of a dot inside the loop were applied. A Keithley multimeter was used for the resistance measurement.

RESULTS AND DISCUSSION

The volume resistivity of the composite in the through-thickness direction was 2.3 Ω cm.

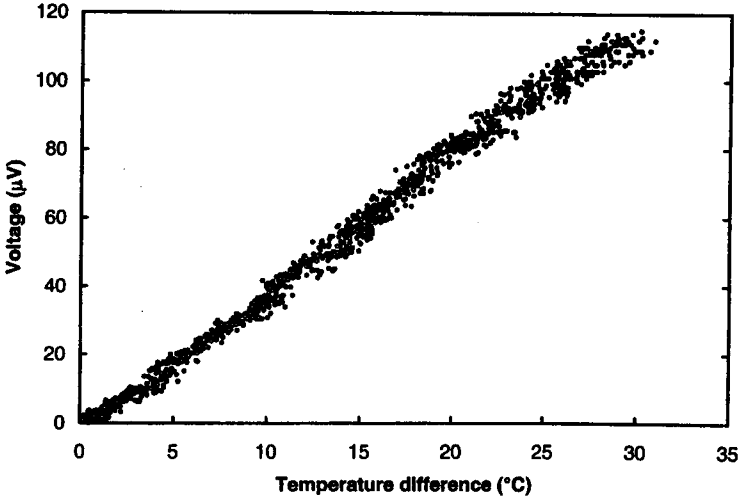

Figure 1. Measured voltage difference vs. temperature difference between the top and bottom surfaces of a composite sheet. ●, during heating; ○, during cooling.

Figure 1 shows the measured voltage difference vs. the temperature difference. The hundreds of data points all fall on a nearly straight line through the origin. The slope of the line gives a thermoelectric power (relative to that of copper) of $+4.0\,\mu V/°C$ ($+3.9\,\mu V/°C$ for the other specimen). This Seebeck coefficient plus the absolute thermoelectric power of copper ($+1.94\,\mu V/°C$ at 300 K) [16] is the absolute thermoelectric power of the sample. The absolute thermoelectric power is thus $+2.1\,\mu V/°C$ ($+2.0\,\mu V/°C$ for the other specimen). This value is roughly comparable to those previously reported for carbons [3–10]. In particular, the value is $-11\,\mu V/°C$ for a carbon–carbon composite laminate (not compliant) in the through-thickness direction [17], as measured at room temperature using the method of this paper.

CONCLUSION

The absolute thermoelectric power of carbon black filled silicone, a compliant material of volume resistivity $2.3\,\Omega\,cm$, is $+2\,\mu V/°C$.

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